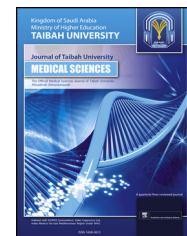




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Experimental Article

An in-vitro assessment of the flow characteristics of different flowable composite systems

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المخلص

أهداف البحث: تهدف هذه الدراسة لمقارنة الخصائص الفيزيائية والميكانيكية للمركب التجاري الجديد القابل للتدفق المنخفض اللزوجة مع غيرها من المركبات القابلة للتدفق المتاحة حاليا في السوق. تمت مقارنة امتصاص الماء ودرجة التحويل باستخدام تنظير الطيف بالأشعة تحت الحمراء.

طرق البحث: تم قياس امتصاص وامتزاز الماء في الماء المقطر واللعب الصناعي باستخدام النقل النوعي، حيث لوحظ الامتصاص والخسارة على فترات زمنية محددة. كما قيس درجة التحويل للروابط المزدوجة للعينات المعالجة وغير المعالجة باستخدام تنظير الطيف بالأشعة تحت الحمراء.

النتائج: أظهرت النتائج زيادة امتصاص الماء في الماء المقطر واللعب الصناعي للمركب، بالمقارنة مع غيره. وكان معامل الانتشار مشابها بشكل عام للامتزاز والامتصاص. وكانت نسبة الذوبان في الماء المقطر أعلى في اللعب الصناعي. أظهرت جميع المواد زيادة في الوزن بعد الامتزاز. أخيرا درجة التحويل كانت متشابهة تقريبا لجميع المركبات التي درست.

الاستنتاجات: إجمالاً هناك فرق في الخصائص الفيزيائية والميكانيكية للمركب المنخفض اللزوجة بالمقارنة بالمركبات ذات التدفق الكبير وتدفق المكان ويتأثر هذا الفرق بوجود هيدروكسي ليثيل ميثاكريلات.

الكلمات المفتاحية: المركبات القابلة للتدفق; التدفق الكبير; تدفق المكان; التدفق المنخفض اللزوجة

Abstract

Objectives: The study aimed to compare the physical and mechanical properties of a new low viscosity commercial flowable composite (Vertise™ Flow) with other flowable composites (Grandio Flow and Premise Flowable) currently available on the market. The water absorption and degree of conversion (using FTIR) were also compared.

Methods: Water absorption and desorption was measured in distilled water and artificial saliva gravimetrically, where the uptake and loss was noted at set time intervals. Degree of conversion of double bonds of uncured and cured samples of composites was measured using the FTIR.

Results: The results showed increased uptake of water in distilled water and artificial saliva for VF, compared to the PF and GF. The diffusion coefficients were generally similar for desorption and absorption. The solubility percentage in distilled water was highest for VF in artificial saliva. All materials showed weight gain after desorption. Lastly, the degree of conversion was found to be almost similar for all the three flowable composites.

Conclusion: The findings of the study conclude that the differences in the physical and mechanical properties of VF compared to GF and PF are influenced by the presence of HEMA.

Keywords: Flowable composite; Grandio flow; Premise flow; Vertise flow

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Introduction

Restorative dentistry is going through a dynamic transition towards adhesive dentistry. A class of resin-composite systems known as 'flowable composites' has become an essential part of the restorative process since their introduction in the mid nineties.¹ These materials were developed in response to a demand from the clinicians for easy handling. They are characterized by having less filler load and greater portion of diluent monomers.² Designed to be less viscous, and so the flowable composites offer a better adaptation to internal walls of the cavity, easier insertion and greater elasticity. Flowability of these materials allows them to be dispensed through injectable dispensers and simplifies easy placement procedures.

Vertise™ Flow (VF) is the flowable composite used in the study that follows. This material was introduced in 1992. It incorporates the Optibond bonding mechanism to dentine by two-fold. Firstly, it binds chemically via the phosphate group of the glycerophosphate dimethacrylate (GPDM) to the calcium of the tooth and secondly micromechanical adhesion by forming a hybrid layer composed of resin impregnating with collagen fibers and dentine smear layer. The University of Leuven, in Belgium, has also proven this adhesion through SEM and TEMs studies. Several investigations have been performed on this material by the American manufacturer Kerr. VF incorporates four types of fillers; the nano-ytterbium that confirms good radiopacity, pre-polymerised fillers which reduce microleakage, improved polishability due to nano particles plus thixotropic properties.

This material was selected for the research due to; its composition which contains poly hydroxyethyl methacrylate (HEMA), a hydrogel, which has the ability of water absorption and increased polymerization exotherm compared with composite matrix monomers.³ As composite restorations are surrounded in an aqueous environment, they tend to absorb water and release un-reacted monomers. These further pose a potential harm in causing allergic reactions and seepage of water which may result in increased bacterial growth and eventually lead to secondary caries. The absorbed water also weakens the filler matrix bond and causes degradation of the material eventually leading to failure of the restoration.⁴ This diffusion controlled water sorption in composites can also cause several time dependent effects which include; hygroscopic expansion, hygroscopic stress leading to cracks in the restoration, weakened mechanical properties, reduction in hardness, glass transition temperature (Ferracane, 2006) and decreased wear resistance.^{5–10}

Finally, the purpose of this research was to compare some of the physical properties of HEMA containing flowable composites to non-HEMA (Grandio Flow and Premise

Flow) containing flowable composites and have a look at the effect of external media (distilled water and artificial saliva) on the three composites.

Materials and Methods

The composites used in this study shown in (Figure 1) are currently marketed in the United Kingdom as low viscosity flowable light cured composites. Vertise™ flow (Kerr Corporation, USA) was compared with another self adhesive flowable composite, Grandio Flow (VOCO, Germany). The control material for the experiment was a conventional flowable composite which uses a separate bonding agent that is Premise Flowable (Kerr Corporation, USA). All the three composite contained nano particle sized fillers along with added other fillers. Composition of all three flowable composites is shown in Table 1.

Two different liquids were used in this study as immersion solutions; distilled water used as the control solvent in this study to observe the water sorption characteristics of the composite samples and artificial saliva. These liquids solvents have been previously used by other investigators and would be a fair representative, of the effects of the liquids in the oral environment on the three composites.⁵

The artificial saliva used was A. S. Saliva Orthana, manufactured by A. S. Pharma, UK. It consists of artificial saliva, mucin, xylitol and fluoride. It is mostly used commercially for the treatment of xerostomia and it is widely used in the Dental Physical Science Unit for transport studies.

Sample preparation

For water absorption/desorption studies, rectangular shaped samples (Figure 2a) measuring 40 mm length, 10 mm width and 1 mm thickness were made using a stainless steel mould. The stainless steel mould was placed on top of a metal plate covered with an acetate sheet. The cavities in the mould were filled with the composite resin. The top surface was covered with another acetate sheet and glass slide. The acetate sheets provided easy removal of the samples from the mould. The samples were then cured with an LED curing light (3MESPE Elipar, Germany) (Figure 2b) for 20 s, using the standard composite cure following manufactures recommendations. Ten over-lapping sections of the specimen were irradiated, respectively, each section for 20 s. The samples were inspected for flaws and were finished by smoothing the edges using a polishing unit (Kent 4 polishing unit manufactured by KemetInt, Ltd, Maidstone, UK). Any samples with visible flaws, porosities or voids were discarded.

Absorption

Six samples of each of the three composite resins were used per immersion solution ($n = 6$). Three measurements were taken on each sample and a mean thickness was recorded using a micrometre screw gauge (accuracy 0.001 mm. Mitutoyo, Japan). The samples were pre-conditioned in a



Figure 1: Flowable composites used in the study.

Table 1: Composition of the flowable composites used in the study.

Material	Resin matrix	Filler	Filler loading/Wt%	Lot no./shade	Manufacturer
Vertise flow	GPDM HEMA	Prepolymerised barium glass filler, colloidal silica, ytterbium fluoride	70	4476058 A2	Kerr Corp, U.S.A
Grandio flow	Bis-GMA TEGDMA HEDMA	2/3rd inorganic fillers	80.2	1206143 A2	VOCO, Cuxhaven, Germany
Premise flow	Ethoxylated bis-phenol-A-dimethacrylate	Trimodal filler system: PPF filler, point-4 filler and 0.02 micron filler	72.5	3791950 A2	Kerr Corp, U.S.A

drying oven at $37 \pm 1^\circ\text{C}$ (P1F120, Camlab, Carbolite, Hope, UK) for 24 h. The immersion solutions (Figure 3) were also placed in an incubator (Thermo Scientific, incubator, IGS 60/100/180) at $37 \pm 1^\circ\text{C}$ for 24 h prior to commencing the absorption studies. An analytical microbalance (Model AE 100, Mettler, Toledo Ltd, Leister UK), weighing to an accuracy of 0.0001 g, was used to weigh each sample prior to immersion in either 100 ml of distilled water or 100 ml

artificial saliva. At noted intervals (refer to Table 2) each sample was removed from the glass jar and blotted on a filter paper (Whatman® filter paper) to remove the excess water.

The sample was weighed and then placed back in the jar. On day one several measurements were taken at intervals to ensure sufficient data for the calculation of the diffusion coefficient. To permit accurate calculations of a timeline

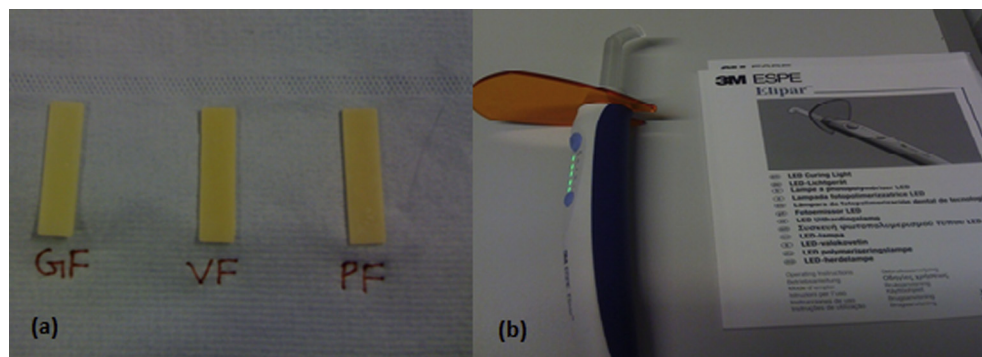


Figure 2: (a) Flowable composites samples in the study. (b) The light curing unit used in this study.

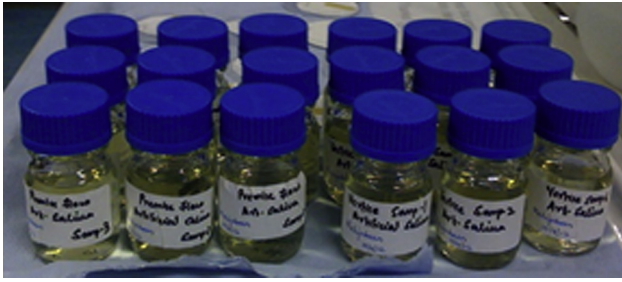


Figure 3: Immersion solutions in screw tight glass jars for water absorption test.

detailed record of the time and date of measurements was kept. The samples in their storage bottles were maintained at 37 °C in the incubator and were only removed for each weight measurements. The water uptake was recorded until there was no further change in weight that is equilibrium had been reached.

Long-term immersion

Eighteen separate bottles were prepared containing artificial saliva and 18 other samples of flowable composites (6 for each composite) were made for a long term immersion study. The samples were pre-conditioned in a drying oven at 37 ± 1 °C (P1F120, Camlab, Carbolite, Hope, UK) for 24 h. The immersion solutions (Figure 3) were also placed in an incubator (Thermo Scientific, incubator, IGS 60/100/180) at 37 ± 1 °C for 24 h prior to commencing the absorption studies. An analytical microbalance (Model AE 100, Mettler, Toledo Ltd, Leister, UK), weighing to an accuracy of 0.0001 g was used to weigh each sample prior to immersion in 100 ml artificial saliva. After 3 months samples were taken out blotted on a filter paper and weighed on a microbalance. The samples were then desorbed using the same method as for the distilled water and artificial saliva short term study.

Desorption

After reaching equilibrium (approximately 5 weeks) the samples were removed from the bottles and placed in an initial desorption drying oven at 37 ± 1 °C (Model P1F120, Camlab, Carbolite, Hope, UK). A similar weighing process to absorption was followed, where samples were individually weighed and readings were recorded at intervals of 5, 10, 20, 40, 60 min intervals on the first day then as for absorption (refer to Table 1). As all the samples were transferred to a

final desorption drying oven kept at 37 ± 1 °C at the end of the second week, kept at 37 ± 1 °C. The water desorption was recorded until there was no significant change in weight.

Diffusion theory and Fick's law

In 1855, according to Adolph Fick, found similarities between diffusion and heat transfer by conduction; he thus proposed that the laws of diffusion were analogous to those governing the laws of heat conductivity. Diffusion can be defined as:

“the process by which matter is transported from one part of a system to another as result of random molecular motions.”¹¹

Fick's mathematical continuum theory governs diffusion in isotropic substances.

Fick's first law describes that the rate of transfer of diffusing substance or fluid through a unit area of material is proportional to the concentration gradient. The rate of diffusion is measured by the diffusion coefficient.¹²

Fick's second law states that the diffusion coefficient can be taken as being independent of concentration, in circumstances for diffusion in dilute solutions. However, the diffusion coefficient is strongly temperature dependant and follows an Arrhenius type relationship. This relates the temperature and the activation energy of a substance to the diffusion coefficient.¹²

Classical theory of diffusion states that during early stages of water uptake (Crank, 1975), the ratio of M_t/M_∞ which represents the ratio of water uptake at time (t) to the uptake at equilibrium is given by:

$$M_t/M_\infty = 2(Dt/\pi L^2)^{1/2} \quad (1)$$

Where D is the diffusion coefficient and $2L$ is the sample thickness.

Similar conditions apply to desorption, except M_t refers to water loss.¹³

Calculating diffusion coefficient

As discussed above, water absorption and desorption are governed by Fick's Law. The ratio M_t/M_∞ which is the ratio of uptake at time (t) to the uptake at equilibrium, and is given by Eq. (1).

If uptake M_t is measured at convenient time intervals until equilibrium M_∞ and M_t/M_∞ is plotted against $(t^{1/2})$, initially the plot should be linear for diffusion controlled uptake/loss. The slope is given by rearranging Eq. (4.1) and gives Eq. (2):

$$S = 2(D/\pi L^2)^{1/2} \quad (2)$$

Hence this can be rearranged to calculate the diffusion coefficient, D

$$D = s^2 \pi (4L^2)/16 \quad (3)$$

Once the samples had equilibrated, they were desorbed. The same equations were applied, with M_t now equals the weight loss (this protocol was adapted from Braden and Clarke).¹⁴

Table 2: Time intervals for weighing of weight change and weight loss of samples.

Period	Weight Intervals
Week 1 (Day1)	0, 5, 10, 20, 40, 60 min, then every hour for 6 h
Week 1 (Day 2)	Two readings
Week 1 (Day 3–5)	Two readings every day
Week 2	Once a day for one week
Week 3	Three readings a week for one week
Week 4	Two readings a week for one week
Week 5	One reading a week for one week

Calculating solubility

The percentage solubility of each sample was calculated using the equation:

$$\% \text{Solubility} = [(W_o - W_d)/W_o] \times 100 \quad (4)$$

Where W_o is the original weight of the sample before immersing in any solution and W_d is the final dehydration weight of the sample. A negative result indicates an overall weight gain. This protocol was adapted from Braden and Davy.¹⁵

Calculating real uptake

To give a value of the total content of water absorbed we calculate the real uptake by using Eq. (5):

$$\text{Max. uptake} + \% \text{ solubility} = \text{Real uptake} \quad (5)$$

Statistical analysis testing was undertaken using an online calculator for *t*-test on Graph Pad (a $p < 0.05$ is deemed significant). Microsoft Excel 2007 was used to plot the data and calculate the mean and standard deviation. The data was analysed statistically, graphically and descriptively.

Results

Water uptake of composites

For each type of composite used in this study (Vertise™ Flow, Grandio Flow and Premise flowable), mean percentage weight change in each solution type, was plotted against square root of time ($t^{1/2}$).

Vertise™ flow

Figure 4 plots percentage weight change against square root of time ($t^{1/2}$) for the 6 samples of VF immersed in artificial saliva, as an example. The data shows the reproducibility between samples.

Average of vertise™ flow, grandio flow and premise flowable in distilled water and artificial saliva

Figure 5 plots the mean percentage weight change against square root of time ($t^{1/2}$) for the three materials in distilled water at 37 °C. We can see that VF shows a steep rise in uptake of water when compared with the other two materials for the same duration of time and this has the highest uptake at 2.5%. PF and GF have similar uptake profiles reaching a maximum of approximately 0.5%. All

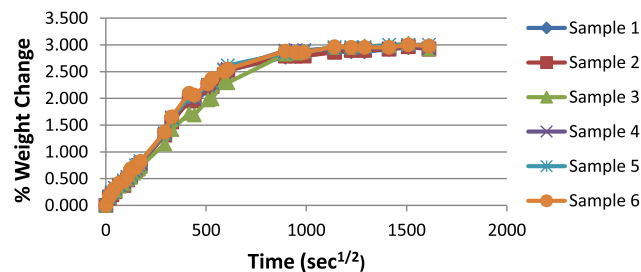


Figure 4: Percentage weight change of VF in Artificial saliva.

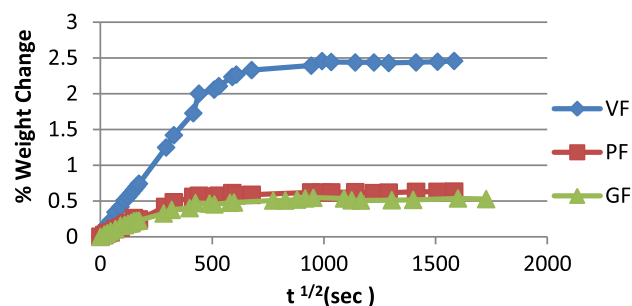


Figure 5: Mean percentage weight uptake of VF, GF and PF in distilled water.

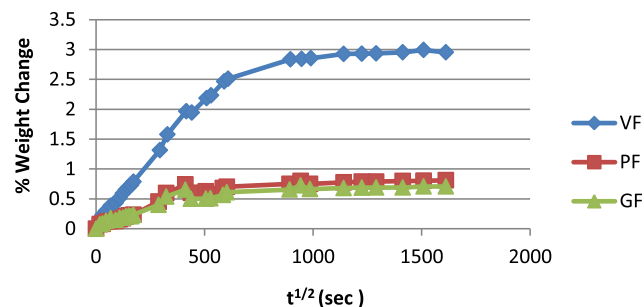


Figure 6: Mean percentage weight uptake of VF, GF and PF in artificial saliva.

materials appear to have reach equilibrium. Overall uptake for VF is significantly ($p = 0.0001$) higher than that of PF or GF ($p \leq 0.05$). While on the other hand uptake of PF and GF are not statistically different ($p = 0.1228$).

Figure 6 plots the mean percentage weight change against square root of time ($t^{1/2}$) for the three materials in artificial saliva at 37 °C. It can be seen that uptake profiles are similar to those in water but maximum uptake is higher ($p = 0.001$). The uptake of all three materials in artificial saliva is significantly different ($p < 0.05$).

Desorption of composites

The same process in generating the water absorption results, were used to generate the water desorption results. All the desorption data was plotted as mean percentage weight loss with square root of time ($t^{1/2}$).

Figure 7 plots the percentage weight loss against square root of time ($t^{1/2}$) for the three materials immersed in distilled water. Overall VF has a significantly ($p = 0.0001$)

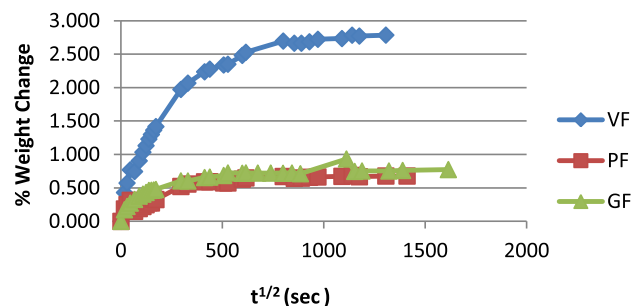


Figure 7: Mean percentage weight loss of VF, GF and PF in distilled water.

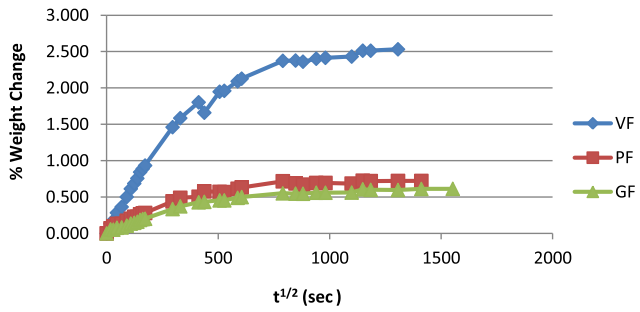


Figure 8: Mean percentage weight loss of VF, GF and PF in Artificial Saliva.

higher loss compared with GF and PF. On the other hand PF and GF are not significantly different ($p = 0.2505$).

Figure 8 plots the percentage weight loss against square root time ($t^{1/2}$) for all the three materials in artificial saliva. It shows that VF has faster rate of loss to a higher loss in weight that PF and GF as in distilled water. The weight loss of all the three materials are significantly different ($p = 0.0001$).

Three months continuous uptake of flowable composite (VF, GF & PF) in artificial saliva

The results of long term (3 months) duration immersion in artificial saliva showed that all the three materials had a significant difference in the water uptake ($p = 0.0001$) as shown in Table 3. Another thing that was noted that the samples of PF and GF took up stains after being immersed in artificial saliva as shown in Figure 9, but on the other hand there was no change of colour on the surface of VF sample, as was also found in studies by the manufacturer (Kerr, 2011).

Figure 10 plots percentage weight loss against square root of time for all the three materials after 3 months continuous immersion in artificial saliva. This graph shows that all the

three materials behave in the same manner as after immersion for a shorter time period however there was a significant ($p = 0.01$) increase in the uptake of water. There also there was a statistical ($p = 0.0001$) difference between weight loss of VF, GF and PF.

Diffusion coefficient for absorption and desorption and solubility %

Using the same data for water/artificial saliva absorption and desorption, the diffusion coefficients, D were calculated. Using data from a representative sample for each material/immersion solution/absorption or desorption combination, M_t/M_∞ was plotted against $t^{1/2}$ (secs $^{1/2}$). The slope of the graph up to $M_t/M_\infty = 0.5$ (to ensure linearity) was determined and D was calculated using Eq. (3).

The percentage solubility of all the samples used for water absorption/desorption studies were calculated, using the Eq. (4). The real uptake was then calculated by combining percentage solubility with maximum percentage absorption as shown in Eq. (5).

Table 3 shows the absorption and desorption diffusion coefficients, maximum percentage uptake, solubility and real uptake for all materials in both immersion solutions.

From Table 3 we can see that the diffusion coefficients of VF in artificial saliva for desorption are much higher compared with distilled water, which is approximately the same for absorption. The solubility of VF is also higher than the other of the materials (PF and GF). However, PF has slower uptake and loss compared with VF and GF. The solubility of PF is also the lowest. GF has almost similar diffusion coefficients in distilled water and artificial saliva for absorption and desorption.

Discussion

The composites chosen for this study are marketed in the UK as low viscosity, self-adhering composites, suitable for

Table 3: Diffusion coefficient of absorption/desorption in different solutions and mean solubility of composites immersed in solvents with standard deviation in parenthesis (negative sign signifies net weight loss).

	Solution	Maximum uptake (SD)	Solubility % (SD)	Real uptake	Diffusion coefficient (m ² sec ⁻¹)
VertiseTM flow	DW	2.46(0.04)	0.43(0.10)	2.89	Absorption 1.47×10^{-13} Desorption 5.26×10^{-13}
VertiseTM flow	AS	2.95(0.03)	-0.31(-0.04)	2.64	Absorption 6.22×10^{-14} Desorption 1.18×10^{-13}
VertiseTM flow	AS/3MNTHS	3.11(0.16)	-0.26(0.03)	2.85	— —
Grandio flow	DW	0.54(0.13)	0.18(0.118)	0.71	Absorption 1.40×10^{-12} Desorption 1.64×10^{-12}
Grandio flow	AS	0.71(0.03)	-0.10(0.02)	0.61	Absorption 1.18×10^{-12} Desorption 8.40×10^{-13}
Grandio flow	AS/3MNTHS	1.10(0.04)	-0.30(0.03)	0.80	— —
Premise flowable	DW	0.63(0.04)	0.06(0.04)	0.69	Absorption 1.83×10^{-14} Desorption 3.18×10^{-14}
Premise flowable	AS	0.81(0.08)	-0.09(0.04)	0.72	Absorption 9.48×10^{-15} Desorption 1.31×10^{-14}
Premise flowable	AS/3MNTHS	0.86(0.13)	0.01(0.34)	0.87	— —

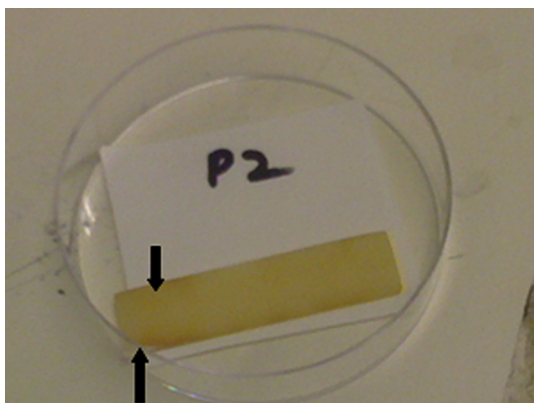


Figure 9: Stains on the sample of PF after long term immersion in artificial saliva.

pits and fissure sealants, repair of marginal defects, liners in deep cavities, class-v restoration and paediatric dentistry.

The company Kerr has formulated a self-adhering, low viscosity nano-hybrid composite known as Vertise™ Flow. It includes the Optibond technology and eliminates the steps of etching/priming/bonding. The bonding mechanism of this material with the tooth structure is a chemical bond achieved via the GPDM phosphate functional groups and calcium ions of the enamel and dentine. According to the company's literature, this material offers high bond strength, high mechanical strength and other physical attributes comparable to other traditional flowable composites. VF is a biocompatible and radiopaque material and bonds well to different substrates including enamel, dentine, porcelain, metals, amalgam and composite.

VF has been a subject of this study here due to its composition containing HEMA and low filler loading (compared with GF and PF). HEMA is a hydrophilic monomer which absorbs water and could potentially decrease the properties of the material.¹⁶

The other materials used in the study were GF and PF. Both these are low viscosity flowable composites. The difference between them and VF is that they do not contain HEMA. These are nano-hybrid composites, and their filler content by weight is 72.5% in PF and 80.2% in GF. They both have good physical and mechanical properties but PF has an added advantage of releasing fluoride and having a higher radiopacity than GF. These materials are in used in this study for comparisons purposes.

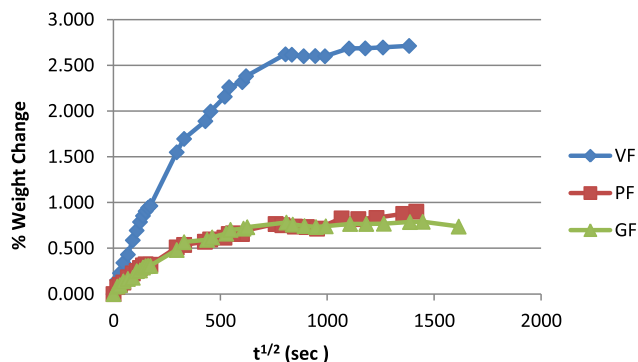


Figure 10: Mean percentage water loss of VF, GF and PF in artificial saliva for 3 months.

Immersion solutions

The immersion solutions chosen for this study were distilled water and artificial saliva, to replicate the daily oral environment and have been used previously by other investigators.^{5,17,18} Distilled water was a control solution in this study and provided a benchmark for the water sorption characteristics. Duplicating the exact properties of human saliva is impossible due to the inconsistent and unstable nature of natural saliva. The use of artificial saliva is essential for well justified and controlled experiments.¹⁹

Water absorption and desorption profile after immersion in distilled water and artificial saliva

According to Rahim et al., flowable composites are becoming more popular and extensively used in dentistry due to their easy handling and mechanical properties.¹² Although surrounded in a wet environment, composites may absorb water or other liquids as saliva which can have an appreciable influence on the degradation of dental composites. Excessive fluid uptake may produce deleterious effects on the structure and function of the resin, as these can reduce the mechanical and physical properties and lead to shorter life of the restoration. On the other hand water ingress may have beneficial effects concerning the expansion of composites, thus compensating for the polymerisation shrinkage, improving the marginal seal and relaxing the stresses set up within the resin matrix during shrinkage.²⁰ These materials after absorbing water, also release unreacted monomers which may stimulate bacterial growth around the restoration and promote allergic reactions.⁵

VF showed the highest amount of water absorption during the period of 35 days immersion period in both distilled water (~2.5% $p = 0.0001$) and artificial saliva (~3%) while GF and PF absorbed ~0.5% water at during this period. The VF results agree with a study by Wei et al., who showed that specimens of VF showed an increased amount of water uptake over 150 days.⁵ Wei et al., concluded that the higher absorption behaviour for VF flowable composites was due to the GPDM monomer introduced by Buonocore et al.^{5,21} However, VF also contains HEMA, a hydrophilic monomer which has been shown to increase water uptake of other methacrylate based material.³ HEMA when polymerised absorbs water to form hydrogel.²² Another reason could be due to the type of filler particles, which in the case of VF are of the nano-hybrid type. Water uptake could also be due to incomplete silanation of filler particles, which will be attacked by the water molecules.²³

In Figures 5 and 6 the absorption profiles for VF in distilled water and artificial saliva were linear for approximately a week before they appeared to equilibrate slowly. PF and GF had very small linear regions and equilibrated after two days. The desorption profiles for the three materials in distilled water/artificial saliva were a reflection of their uptake processes (Figures 7 and 8). In this study it was anticipated (and has been confirmed) that VF would show an increase in water uptake compared with GF and PF ($p = 0.0001$). However, the manufacturer has neglected to put in water uptake data, for this material in their literature, whereas data is available for GF and PF.

Table 4: Summary of diffusion coefficient for absorption and desorption.

Medium	$D \text{ m}^2\text{sec}^{-1}/(\text{Max.uptake } \%)$	$D \text{ m}^2\text{sec}^{-1}/(\text{Max.uptake } \%)$	$D \text{ m}^2\text{sec}^{-1}/(\text{Max.uptake } \%)$
	VertiseTM flow	Grandio flow	Premise flowable
Distilled water uptake	1.47×10^{-13} (2.46)	1.40×10^{-12} (0.54)	1.83×10^{-14} (0.63)
Distilled water loss	5.26×10^{-13}	1.64×10^{-12}	3.18×10^{-14}
Artificial saliva uptake	6.22×10^{-14} (2.95)	1.18×10^{-12} (0.71)	9.48×10^{-15} (0.81)
Artificial saliva loss	1.18×10^{-13}	8.40×10^{-13}	1.31×10^{-14}

Solubility of the composites

Water plays an important role in the oral environment as the solvent of aqueous solutions and the ingested liquid. Dental composites continuously interact with water after being applied intra orally. Thus it's logical that they will absorb water and release unreacted monomers, initiators, catalyst, stabilizers or soluble materials.⁵ Degradation and erosion of dental composite will occur as a result of incomplete polymerisation and influence of oral fluids.^{24,25} Degradation of composites depend on various factors like composition of monomers, degree of conversion and environmental impact.²⁶ Residual monomers are reported to be the major components being released, within the first 7days of placement of cured dental composites.²⁷

From Table 3 it can be seen that all three flowable composites resulted in low solubilities in distilled water, ranging from 0.06% for PF to 0.43% for VF. In artificial saliva all three materials showed a very small gain in weight (negative solubility). This could be due to either water being bound to groups within the material, or some interaction between components of artificial saliva and the composite. Similar negative solubility values were found by Teh, in water absorption studies of nano/microhybrid composites.¹⁰

This study found that there were statistical differences ($p = 0.0001$) in solubility between VF, GF and PF immersed in both solutions indicating that the amount of elution from all the composites was different. To identify leachants from the tests materials further test and investigations are needed. Braden, recommended HPLC (high performance liquid chromatography) could be used to identify such small amounts of leachant material.²⁹

Diffusion coefficient of the composites

Dental composite resins absorb water in the oral cavity.^{5,20} Asaoka and Hirano (2003) suggested that the diffusion coefficient is important in determining the time-dependant mechanical properties and the time-dependant hydroscopic expansion of resins for clinical use. Diffusion coefficient, which represents the speed/rate of water diffusing in the resin matrix, could be related to composite degradation. The higher the diffusion coefficient, the faster would be the degradation occurring in composite. The absorption phenomenon in composites is a diffusion controlled process.¹⁸ High diffusion coefficient molecules (e.g. $< \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$) mean that the diffusant is entering faster than for low molecules (e.g. $> \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$).³⁰

From Table 4 it is evident that there was little variation in diffusion coefficient for uptake and loss processes in both

immersion solutions (distilled water and artificial saliva), for all three flowable composites. This indicates the diffusion coefficient is concentration dependant. Earlier studies on composites and acrylics reported that the diffusion coefficient for desorption were higher than the absorption processes indicating that the diffusion coefficient were concentration dependant.^{28,31}

From Table 4 it can be seen that GF (over the 35 day study) had the lowest uptake in distilled water and artificial saliva ($p = 0.0001$) compared with GF and PF. There also appears to be very little difference in diffusion coefficients in uptake and loss for distilled water and artificial saliva for this material, and they were higher than VF and PF. GF and PF showed similar profiles for both uptake and loss processes. Wei et al. found that VF at 42nd day of immersion in distilled water, exhibited significantly higher water absorption solubility and diffusion coefficient ($5.23 \times 10^{-9} \text{ cm}^2/\text{s}$) than the other flowable composites.⁵

Conclusions

VF has statistically shown higher water uptake and higher loss than GF and PF after immersing in distilled water and artificial saliva. Diffusion coefficients were similar for both absorption and desorption in distilled water and artificial saliva for all the three materials. VF had the highest solubility compared with GF and PF in distilled water, but were in the order of $\text{GF} > \text{VF} > \text{PF}$. All three flowable composites demonstrated weight gain in artificial saliva. There was no difference in degree of conversion in the three materials.

Authors contribution

Acquisition of data of data was particularly done by Dr. Tayyaba Fatima whereas further analysis and interpretation of data was done in collaboration of both the author's. Further being Corresponding Author drafting and critical revision was done by Dr. Zeeshan Qamar.

Conflict of interest

The authors have no conflict of interest to declare.

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